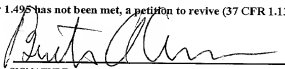


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<b>TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371</b>		<b>ATTORNEY'S DOCKET NUMBER</b>  0679/00188	
<b>INTERNATIONAL APPLICATION NO.</b> PCT/IT99/00195		<b>INTERNATIONAL FILING DATE</b> 1 July 1999	<b>U.S. APPLICATION NO. (If known, give 37 CFR 2.5)</b> <b>09/529726</b> <b>412 Rec'd PCT/PTO 19 APR 2000</b>
<b>PRIORITY DATE CLAIMED</b>			
<b>TITLE OF INVENTION</b> MEMBRANE-SEPARATED, BIPOLAR MULTICELL ELECTROCHEMICAL REACTOR			
<b>APPLICANT(S) FOR DO/EO/US</b>  Broman, Barry Michael, Zocchi, Andrea			
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:			
<ul style="list-style-type: none"><li>1. <input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371</li><li>2. <input type="checkbox"/> This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. § 371.</li><li>3. <input checked="" type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).</li><li>4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.</li><li>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2))<ul style="list-style-type: none"><li>a. <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau).</li><li>b. <input checked="" type="checkbox"/> has been transmitted by the International Bureau.</li><li>c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).</li></ul></li><li>6. <input checked="" type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)).</li><li>7. <input type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))<ul style="list-style-type: none"><li>a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).</li><li>b. <input type="checkbox"/> have been transmitted by the International Bureau.</li><li>c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</li><li>d. <input type="checkbox"/> have not been made and will not be made.</li></ul></li><li>8. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</li><li>9. <input type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).</li><li>10. <input type="checkbox"/> A translation of the Annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).</li></ul>			
<b>Items 11. to 16. below concern other document(s) or information included:</b>			
<ul style="list-style-type: none"><li>11. <input checked="" type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.</li><li>12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</li><li>13. <input type="checkbox"/> A FIRST preliminary amendment. <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment.</li><li>14. <input type="checkbox"/> A substitute specification.</li><li>15. <input type="checkbox"/> A change of power of attorney and/or address letter</li><li>16. <input checked="" type="checkbox"/> Other items or information: International Search Report</li></ul>			

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U.S. APPLICATION NO. (If known, see 37 CFR 1.5) <b>09/529726</b>		INTERNATIONAL APPLICATION NO. PCT/IT99/00195		ATTORNEY'S DOCKET NUMBER 0679/00188	
x The following fees are submitted:				CALCULATIONS	PTO USE ONLY
<b>Basic National Fee (37 CFR 1.492(a)(1)-(5)):</b> Search Report has been prepared by the EPO or JPO ..... \$840.00 International preliminary examination fee paid to USPTO (37 CFR 1.482) ..... \$670.00 No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)) ..... \$760.00 Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... \$970.00 International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4) ..... \$96.00					
<b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>				\$840	
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				\$	
Claims	Number Filed	Number Extra	Rate		
Total Claims	6- 20 =	0	X \$18.00	\$	
Independent Claims	1- 3 =	0	X \$78.00	\$	
Multiple dependent claim(s) (if applicable)			+ \$260.00	\$	
<b>TOTAL OF ABOVE CALCULATIONS =</b>				\$840	
Reduction by 1/2 for filing by small entity, if applicable. Verified Small Entity statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28)				\$	
<b>SUBTOTAL =</b>				\$840	
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				\$	
<b>TOTAL NATIONAL FEE =</b>				\$840	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property +				\$	
<b>TOTAL FEES ENCLOSED =</b>				\$840	
				Amount to be:	
				refunded	\$
				charged	\$
a. x A check in the amount of \$840 to cover the above fees is enclosed. b. <input type="checkbox"/> Please charge my Deposit Account No. <b>22-0185</b> in the amount of \$_____ to cover the above fees. A duplicate copy of this sheet is enclosed. c. x The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <b>22-0185</b> . A duplicate copy of this sheet is enclosed.					
NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status SEND ALL CORRESPONDENCE TO: <b>Pollock, Vande Sande &amp; Amernick, R.L.L.P.</b> 1990 M Street, N.W., Suite 800 Washington, DC 20036-3425					
				SIGNATURE  Burton A. Amernick NAME 24.852 REGISTRATION NUMBER	

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Applicant/Patentee: Broman et al  
Serial/Patent No.: To be assigned  
Filed on/Issued on: April 19, 2000  
For: Membrane-Separated, Bipolar Multicell Electrochemical Reactor



**VERIFIED STATEMENT (DECLARATION) CLAIMING SMALL ENTITY  
STATUS (37 CFR § 1.9(f) AND § 1.27(b)) - SMALL BUSINESS CONCERN**

I hereby declare that I am:

- ☐ The owner of the small business concern identified below:  
☒ An official of the small business concern empowered to act on behalf of the concern identified below:

NAME OF CONCERN: CHEMIECO s.r.l.

ADDRESS OF CONCERN: Erta Canina, 68, 50125 Firenze, Italy

I hereby declare that the above-identified small business concern qualifies as a small business concern as defined in 13 CFR § 121.8-18, and reproduced in 37 CFR § 1.9(d), for purposes of paying reduced fees under 35 USC § 41(a) and (b), in that the number of employees of the concern, including those of its affiliates, does not exceed 500 persons. For purposes of this statement: (1) the number of employees of the business concern is the average over the previous fiscal year of the concern of the persons employed on a full-time, part-time or temporary basis during each of the pay periods of the fiscal year, and (2) concerns are affiliates of each other when either, directly or indirectly, one concern controls or has the power to control the other, or a third party or parties controls or has the power to control both.

I hereby declare that the rights under contract or law have been conveyed to and remain with the small business concern identified above with regard to the invention entitled Membrane-Separated, Bipolar Multicell Electrochemical Reactor by inventor(s) Barry M. Broman and Andrea Zocchi described in:

- ☐ The specification filed herewith  
☒ U.S. Application Serial No. , filed April 19, 2000  
☐ U.S. Patent No. , issued

If the rights held by the above-identified small business concern are not exclusive, each individual, concern or organization having rights to the invention is listed below\* and no rights to the invention are held by any person, other than the inventor, who could not qualify as an independent inventor under 37 CFR § 1.9(c), if that person made the invention, or by any concern which would not qualify as a small business concern under 37 CFR § 1.9(d) or a non-profit organization under 37 CFR § 1.9(e).

FULL NAME  
ADDRESS

☐ Individual ☒ Small Business ☐ Non-Profit

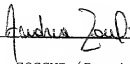
\*NOTE: Separate Verified Statements are required from each named person, concern or organization having rights to the invention averring to their status as small entities (37 CFR § 1.27).

I acknowledge the duty to file, in this application or patent, notification of any change in status resulting in loss of entitlement to small entity status prior to paying, or at the time of paying, the earliest of the Issue Fee or any maintenance fee due after the date on which status as a small entity is no longer appropriate (37 CFR § 1.28(b)).

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 18 USC § 1001, and that such willful false statements may jeopardize the validity of the application, any patent issuing thereon, or any patent to which this Verified Statement is directed.

CHEMIECO s.r.l.

By:



Title: Andrea ZOCCHI (President)

Date: June 8, 2000

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MEMBRANE-SEPARATED, BIPOLAR MULTICELL

ELECTROCHEMICAL REACTOR

BACKGROUND OF THE INVENTION

1. FIELD OF THE INVENTION

- 5 This invention relates to electrochemical reactors for conducting reduction and oxidation reactions in respective positive and negative liquid electrolytes, without gas evolution at the electrodes. More specifically the invention relates to a membrane-separated, bipolar multicell electrochemical reactor for implementing a redox flow battery system.

10 2. DESCRIPTION OF RELATED ART

Redox flow battery systems are increasingly attracting interest as efficient energy conversion systems. Among redox couple candidates, the all vanadium redox system being one of the most preferred.

- 15 Structurally, the electrochemical reactors that have been proposed for redox flow battery systems, have been derived from the electrochemical reactor structures developed for general electrolysis processes, the only adaptation having concerned the materials employed as electrodes.

- 20 Generally, the electrochemical reactors used as redox batteries are conventionally composed of a stack of bipolar plate electrode elements separated by ion exchange membranes, defining a positive electrolyte flow chamber on one side of each membrane and a negative electrolyte flow chamber on the opposite side thereof. The stack of bipolar elements is assembled together in a filter-pass arrangement between two end electrode elements.

- 25 Commonly, The elements have a frame provided with co-ordinated through holes forming inlet and outlet manifolds for the two electrolytes that are circulated in a parallel mode through the positive electrolyte flow chambers and the negative electrolyte flow chambers, respectively.

The elements are conventionally mounted and operated in a vertical position.

The assembling of a large number of bipolar elements in electrical series as required in redox batteries to reach an adequate voltage at the two ends of the battery, the positioning of innumerable gaskets for sealing the outer perimeter of each electrolyte flow chamber and the perimeter of the distinct through holes of the frames for defining the inlet and outlet manifolds for the two electrolytes and the final tightening of the filter press assembly by tie rods compressing the two end elements over the stack, are extremely delicate and time consuming operations that require particularly skilled technicians.

- 10 The parallel flow of the two electrolytes through the respective flow chambers poses serious problems in terms of minimization of so-called stray or by-pass electric currents in uninterrupted liquid veins of electrolyte, due to the fact that the electrolyte present in the manifolds offer innumerable paths for these by-pass or stray currents, driven by mutual voltage differences existing among the various
- 15 bipolar elements functioning in electrical series between the two end electrodes on which the full battery voltage difference insists. By-pass or stray currents decrement the energy efficiency of the conversion system, but more seriously they cause severe corrosion phenomena on conductive parts (e.g.: carbon) because of extremely high half-cell voltages at the conductor surface.
- 20 On the other hand, the redox system require nonnegligible electrolyte flow rates through the flow chambers of the reactor in order to maintain optimal half-cell reactions conditions at the electrodes and this requirement may imply the necessity of operating the bipolar electrochemical reactor at relatively high positive pressure.
- 25 Differently from conventional electrochemical processing, redox flow battery systems are intended also for uses on nonpolluting vehicles and power/weight ratio is an important parameter.

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### SUMMARY OF THE INVENTION

A main objective of this invention is to provide a membrane-separated bipolar multicell electrochemical reactor for half-cell reduction and oxidation reactions in respective positive and negative electrolytes, without gas evolution, with an architecture that makes it more easily assemblable by allowing to stack fully pre-assembled elements horizontally, one on top of the other, and suitable to be operated in the same horizontal orientation of the bipolar elements.

According to a fundamental aspect of the novel architecture of the invention, the multicell assembly is constituted by alternately stacking two types of pre-assembled elements, one being a bipolar electrode holding subassembly and the other being a membrane holding subassembly.

Of course, the alternate stack of elements is piled over a bottom end element and the stack is terminated by placing over the last membrane holding element a top end electrode element. The two end electrode elements are then compressed over the stack by tightening a plurality of tie rods, conventionally arranged around the perimeter of the stacked elements, according to a common practice in tightening a filter-press stack in a hydraulically sealed manner, by virtue of the gaskets operatively installed between the coupling faces of the frames of the various elements.

According to an essential aspect of the architecture of the invention, each bipolar plate electrode holding element and each ion exchange membrane separator holding element includes a substantially similar rectangular frame piece, made of an electrically nonconductive and chemically resistant material, typically of molded plastic material, having on its upper (assembly) face grooves for receiving O-ring type gasket means, and having through holes and recesses in coordinated locations disposed along two opposite sides of the rectangular frame forming, upon completion of the assembling, ducts for the separate circulation of the

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negative electrolyte and of the positive electrolyte through all the negative electrolyte flow chambers and all positive electrolyte flow chambers, respectively, in cascade. The negative electrolyte enters along a first side of a negative electrolyte flow chamber, flows through the chamber toward the opposite or  
5 second side thereof, exits the chamber, flows through the coordinated holes through the frame holding the electrode and through the frame holding the next membrane separator, reaching the level of the next negative electrolyte flow chamber and enters it from the same second side through which it exited from the previous negative electrolyte flow chamber and exits this next negative electrolyte  
10 flow chamber from the same first side it entered the previous negative electrolyte flow chamber, to flow through coordinated holes through the next pair of frames to the level of the next negative electrolyte flow chamber and so forth. The same flow path is arranged also for the positive electrolyte, either in a "countercurrent" or in an "equicurrent" mode through the battery.

15 In practice, the bipolar electrochemical reactor does not have inlet and outlet manifolds for the two electrolytes, on the contrary, the electrolytes flow through the respective flow chambers in a zigzag path, that is essentially in hydraulic series or cascade mode instead than in hydraulic parallel mode.

20 In this way, by-pass current may only be "driven" by a voltage difference of about one-cell voltage and becomes practically negligible and above all it does not cause any corrosion on conductive parts.

The two types of pre-assembled elements are coordinately "keyed" so as to prevent any error in correctly stacking them alternately one over the other and with a correct orientation and perfect mutual alignment to compose the bipolar  
25 battery.

Apart from the suitably shaped keying pins and sockets and the position of the through holes and of the slotted portions of communication with the flow

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chambers, the molded plastic frames are substantially identical for both types of elements.

Essentially, each frame has an inner flange portion, recessed from the bottom (assembly) face of the frame, that is the opposite face to the one that is provided  
5 with the grooves for accommodating O-ring gaskets around pass-through electrolyte ducting holes and around the outer seal perimeter of the chamber.

During the pre-assembling of the two types of elements, on this inner flange portion is accommodated a relatively narrow edge perimetral portion of either a bipolar plate electrode or of an ion exchange membrane separator.

10 The two types of frames may conveniently be made of a different color or tonality for an easy recognition of which is destined to accommodate a bipolar plate electrode or an ion exchange membrane.

The face of the flange portion on which the ion exchange membrane separator or the bipolar plate electrode is set, has a plurality of orderly spaced retention pins  
15 that project out of the surface of the flange.

The plate electrodes and the ion exchange membrane separators are distinctively from each other, provided with coordinated through holes into which the retention pins of the respective frame piece pass.

A retention counterflange of an electrically nonconductive and chemical resistant  
20 material, typically of the same material and color of the respective type of frame, has also a number of holes coordinated with the positions of the retention pins and is functionally mounted over the perimetral portion of the plate electrode or of the membrane separator, whichever belongs to the particular type of frame, placed on the recessed flange portion of the frame.

25 The retention counter flange is fixed in position by flattening with a hot tool the protruding portions of the retention pins thus permanently fixing the bipolar plate

electrode or the ion exchange membrane separator in the central window of the respective frame.

The rectangular shape of the frame windows into which membranes and bipolar plate electrodes, cut or made to size, are fitted, minimizes any waste of valuable  
5 membrane and bipolar plate material. A tight fitting of the counterflange effectively seals the contour of the membrane or bipolar plate preventing electrolyte intermixing reducing the number of gaskets. Optionally spacer gaskets may be used when necessary, for example to mount a particularly thin membrane.

This arrangement permits to overturn the so pre-assembled elements without the  
10 risk that the fitted bipolar plate electrode or ion exchange membrane separator may fall off and therefore permits to easily dispose the O-ring gaskets on the opposite (upper) face of the frame suitably provided with accommodating grooves.

Each pre-assembled element of a first type, may be overturned, placed on top of  
15 the stack and O-ring gaskets may be placed in the respective grooves before placing the next pre-assembled element of the other type on top, ready to receive the O-ring gaskets thereon and another pre-assembled element of the first type, and so on until completing the stack.

The keying pins and sockets, besides obliging a correct alternate stacking and  
20 orientation of bipolar electrodes and membrane separators pre-assembled elements, impose also a correct orientation on the plane of the elements such that the though holes and slotted portions coordinate with each other realizing the zigzag serial flow path of the two electrolytes.

Preferably the battery stack is made up of an integer multiple number of blocks  
25 each constituted by four alternately coupled elements: two membrane elements and two bipolar elements the battery having an even number of cells. In this way each electrolyte will enter and exit the battery from the same side.

Often and most preferably, the electrode consists of a porous fabric or mat of carbon fibers in electrical continuity with a similar electrode structure on the opposite face of the conductive bipolar plate electrode for providing substantially three-dimensional electrode structures having a large active area, that extends for a considerable portion into the depth of the relative electrolyte flow chamber.

This arrangement, dictated by the need to increase the allowable rate of half-cell reaction process at the electrode, contrasts with the need to minimize the power absorbed by the motors that drive the pumps of the two electrolytes to flow them through the plurality of respective flow chambers at an adequate flow rate.

This problem is aggravated when passing from a parallel flow of the electrolyte through all the respective flow chambers from a common inlet manifold to common outlet manifold to a cascade flow from one chamber to the next and so forth from one end to the opposite end of the stack.

Although this cascade flow mode is extremely effective in eliminating any corrosion problems due to by-pass currents, it necessarily implies an augmented pressure drop through the battery of the two electrolytes.

According to an important optional feature of the battery architecture of the invention, useful in case of use of porous three-dimensional electrodes extending from the impervious electrically conductive bipolar plate, this increase of pressure drop due to the use of a series or cascaded flow path of the electrolytes through the respective pluralities of flow chambers when using porous mat electrodes encroaching into the flow chamber is practically eliminated while reducing or even eliminating any residual gap or unobstructed flow space between the porous electrode and the ion exchange membrane separator, which, may even be placed in contact with each other to minimize ohmic losses in the liquid electrolyte.

These apparently contradictory conditions are indeed accomplished according to the present invention by defining (cutting) in the porous electrode two orders of

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parallel flow channels, all the parallel spaced channels of each order extend from a common orthogonal base channel formed along the respective inlet or outlet side of the chamber and terminate short of reaching the base channel of the other order. Each order defines a comb-shaped flow distributing channelwork the parallel fingers of which interleave with the finger channels of the other order.

Practically, one comb-shaped channelwork has its base or manifold channel running along a side of the chamber communicating with the inlet duct of the electrolyte into the chamber while the other specular comb-shaped channelwork has its parallel finger channels interleaved with the parallel finger channels of the first channelwork and has its base or manifold channel running along the opposite side of the chamber communicating with the outlet electrolyte duct.

The interleaved finger flow channels or one order run parallel to each other, each terminating short of the manifold base channel of the other order of interleaved parallel finger channels. Therefore, each inlet or "source" flow channel is separated from the two adjacent outlet or "drain" flow channels by a strip of a certain width of the three-dimensional porous electrode material, separating the parallel channels that may be eventually cut in it.

The interleaved orders of inlet and outlet electrolyte flow channels evenly distribute the electrolyte with a reduced pressure drop uniformly throughout the electrode area of the flow chamber, providing flow distribution channels throughout the mass of the three-dimensional porous electrode.

The pressure drop may be pre-arranged within a certain margin, by knowing the specific pressure drop of the electrolyte through the three-dimensional porous electrode material at a given flow rate and by designing the two orders of interleaved "source" and "drain" flow channels with an appropriate distance of separation from one another.

Besides outstandingly reducing the pressure drop suffered by the electrolyte

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flowing through the respective flow chambers in series from one end to the opposite end of the battery, this arrangement of interleaved "source" and "drain" channels, suitably cut through the thickness of the three-dimensional porous electrode, is found to outstandingly enhance the electrochemical performance of the battery because of a far more evenly distributed current density over the entire cell area of the battery.

The invention is more clearly defined in the annexed claims.

#### BRIEF DESCRIPTION OF THE DRAWINGS

The different features and related advantages of the novel electrochemical battery architecture will become even more evident through the following description of several preferred embodiments and by referring to the attached drawings, wherein:

**Figure 1** is an exploded view of a membrane element and a bipolar electrode element according to a preferred embodiment;

**Figure 2** is a fragmentary detail view of an upturned membrane element detailing the way the membrane is fitted into the rectangular window of the frame;

**Figure 3** is a fragmentary detail view of an upturned bipolar electrode element detailing the way the membrane is fitted into the rectangular window of the frame;

**Figure 4** is a plan view of a bipolar plate electrode element using electrode structures in the form of a carbon felt or fabric, according to a preferred embodiment of the invention;

**Figure 5** is a fragmentary delay cross section of the bipolar plate electrode of Figures 1 and 3;

**Figure 6** is a cross sectional three-dimensional view of a completely assembled battery stack.

#### DESCRIPTION OF SEVERAL EMBODIMENTS OF THE INVENTION

Figures 1, 2, 3, 4 and 5 illustrate a battery realized according to a preferred

embodiment of the present invention.

The exploded view of Fig. 1, representing the two types of the pre-assembled elements, namely a membrane element and a bipolar electrode plate element, provides an explicative overview of the architecture of the invention, according to a preferred embodiment.

The frame portion 1M of membrane element and the frame portion 1E of a bipolar electrode element are under many aspects substantially similar. Both may be of a molded plastic material such as for example polypropylene, different molds being employed for the two types of frames 1E and 1M, for implementing suitably different keying pins and sockets.

According to a most preferred embodiment, the molded plastic frames 1M and 1E define a rectangular inner window, however, the outer perimeter of the frame is not rectangular but has a peculiarly curved shape, determined by the fact that each side of the frame has a convex outer edge, making the width of the cross section of the frame larger near the central portion of each side than near to corners. This particular shape of the frame optimizes weight versus structural strength, in consideration of the electrolyte pressure withstanding requisites. Indeed, the completed battery assembly is hydraulically tested to withstand a maximum electrolyte pressure of up to 5 atmospheres. The convex shape of the sides of the frame body has been found to best respond to the flexural strength requisite by reducing stress concentrations and achieving a most efficient weight/pressure resistance ratio.

In the exploded view of Fig. 1, the flow paths of the positive electrolyte and of the negative electrolyte through the respective positive electrode and negative electrode flow chambers are schematically traced by flow lines depicting the respective positive (+) and negative (-) signs.

The electrolyte flow paths are shown, according to stacking orientation of the

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elements of the battery starting from a horizontally disposed bottom terminal element consisting of a positive end electrode.

In the figure, the lower element is a membrane element showing a rectangularly cut membrane 2 which may be either a cation exchange membrane such as  
5 sulphonated polyethylene membrane or a polystyrene sulphoric acid membrane or similar chemically resistant membrane or an anion exchange membrane.

The membrane 2, cut to size, has also a number of holes 3 punched along its perimetral portion at pre-ordinated positions. Similarly, a fixing counterflange 4, generally of the same material of the frame piece 1M, has holes at pre-ordered  
10 positions, geometrically coinciding with the positions of the holes 3 of the membrane 2.

By looking at Fig. 2, a detail enlarged view of the membrane element of Fig. 1, overturned by 180°, it is shown how the membrane 2 and the fixing counterflange 4 are accommodated over the recessed inner flange portion 6 of the frame 1M, the  
15 coordinated holes 3 and 5 of the membrane and of the retaining counterflange 4 respectively fitting on the array of molded retention pins 7 present on the face of the flange portion 6.

Spacer rubber gaskets may be disposed on the flange portion 6 before installing the membrane 2 and a second rubber gasket may be interposed between the  
20 membrane 2 and the fixing counterflange 4 to adjust to the required depth of the respective flow chambers on the opposite sides of the membrane 2 and/or to prevent leakages of the negative electrolyte into the positive electrolyte or viceversa by possible pressure differences in the two flow chambers on the opposite sides of the separating membrane 2.

25 Alternatively, a suitable sealant such as for example a silicone gel sealant may be used in pre-assembling the element to ensure a leakproof assembly.

By appropriate implements, the retaining counterflange 4 is eventually pressed over the membrane and the ends of the retention pins 7 protruding out of the holes 5 of the counterflange 4 are heat-riveted by the use of a heated tool, causing the "molding" of the molten tip of the pins 7 into the swaged holes 5 of the fixing flange 4.

Upon "riveting" the retention pins 7, the element is completely pre-assembled and may be stacked over a last-mounted bipolar electrode element in an overturned condition, without any risk for the pre-installed membrane to fall off.

Moreover, the upper face of the molded frame piece 1M of the membrane element may readily receive, if not already installed, a perimetral seal O-ring gasket 8 and the two negative (or positive) electrolyte ducts O-rings, 9 and 9'.

The upper face of the frame piece 1M of the membrane element has at least two, preferably four, keying pins 10, which, in the example, have an oblong cross section, while on the lower face, partially visible in the detail view of Fig. 2, two or preferably four pin sockets 11 are present, which in the example shown, have a circular cross section.

In the example shown in the figures, the tie rods that complete the battery assembly pass through the holes 12, formed ordered in the perimetral portion of all the frames and of the two end elements, external to the perimetral seal O-ring gasket 8.

The upper group of the exploded view of Fig. 1, depicts a bipolar electrode element.

According to a most preferred embodiment of the battery of the invention, the bipolar plate electrode is an electrically conductive composite that includes a central conductive plate 13, typically a glassy carbon plate with a thickness in the order of 1 to 3 or more millimeters, having bonded over its opposite faces a



carbon felt electrode structure 14 of a thickness (depth) that may generally be comprised between 1 and 5 or more millimeters.

The felt electrodes 14 are bonded in electrical continuity to the conductive septum 13 and have a relatively open structure such to be readily permeated by the electrolyte flown through the electrode chamber.

The bipolar plate electrode composite, 13-14, is mounted in a respective frame piece 1E of a molded thermoplastic, typically of the same type with which are produced the frame pieces 1E of the membrane elements.

The pre-assembling of the bipolar electrode element is totally similar to the pre-assembling of a membrane element.

A different disposition of the coordinated plurality of holes 16 in the perimetral portion of the electrically conductive carbon plate septum 13 and of the holes 17 in the retention counterflange 15, and of course also of the retention pins 18 (Fig. 3) on the recessed inner flange portion 19 of the frame 1E, will prevent any possibility of error in pre-assembling the two types of stackable elements.

Fig. 3 is an enlarged detail view of the bipolar electrode element of Fig. 1, overturned by 180°.

Differently from the frame piece 1M of the membrane element, the frame piece 1E of the bipolar electrode element has, on the upper face keying pins 20 (Fig. 1) having a circular cross section of a diameter appropriate to fit into the circular sockets 11 present on the underside of the frame piece 1M of a membrane element. As visible in the upturned view of Fig. 3, on the upper side of the molded frame piece 1E of the bipolar electrode element (Fig. 1), there are sockets 21 with an oblong cross section suitable to accommodate the keying pins 10 present on the upper side of the molded frame piece 1M of the membrane element.

Similarly to the frame piece 1M of the membrane elements, also the frame piece 1E of the bipolar electrode element has on its upper face grooves for accommodating a perimetral sealing O-ring 22 and two positive (or negative) electrolyte ducts O-rings 23 and 23'.

- 5 According to a most preferred embodiment depicted in the drawings, the porous carbon mat electrodes 14 have two distinct orders or channelworks of parallel and mutually interleaved electrolyte distributing channels oriented along the same direction of the electrolyte flow through the respective electrolyte flow chamber, starting from the inlet recessed slots 24 and 24' and 25 and 25' for the positive  
10 and negative electrolytes, respectively, to the outlet recessed slots 26 and 26' and 27 and 27', functionally present on opposite sides of the chamber.

Of course, though in the example two ducts and relative slots are shown, depending on the side of the cells there may be only one duct and slot or any number of ducts and slots for a better distribution and/or for reducing hydraulic  
15 pressure drops.

As depicted in Figures 1, 3, 4 and 5, a first "comb-shaped" channelwork has the finger channels s1, s2, s3, ..., sn, extending parallel to each other from a base or manifold channel S defined along the side of the electrolyte flow chamber through which the electrolyte enters the chamber through the inlet slots,  
20 connecting with the respective electrolyte ducts, and end short from reaching the corresponding manifold or base channel D defined along the opposite side of the flow chamber where the electrolytes exits the chamber through the outlet slots communicating with the respective electrolyte ducts.

Similarly, the second comb-shaped channelwork has finger channels d1, d2, d3, ...  
25 dn, extending parallel to each other from their base or manifold channel D and they are interleaved with the finger channels s1, s2, s3, ..., of the first comb-shaped channelwork. The finger channels d1, d2, d3, ... of the second comb-

shaped channelwork, terminate short of reaching of the base or manifolding channel S of the first channelwork.

The two interleaved orders of channels, s1, s2, s3, ..., and d1, d2, d3, ..., constitute an electrolyte distributing (source) channelwork and an electrolyte draining (drain) channelwork.

As may be observed in the plan view of Fig. 4 and in Figures 1 and 3, the respective base or manifolding channels S and D for the incoming electrolyte and for the electrolyte exiting the flow chamber, respectively, are defined by having the felt electrode 14 ending at a certain distance from the side wall of the flow chamber so that the electrolyte entering the flow chamber through the inlet slots 24 and 24' (Fig. 3) or 25 and 25' (Figures 1 and 4) is able to distribute itself in this base or manifolding inlet channel S and from there to evenly distribute itself along the source finger channels s1, s2, s3, ... .

The drain finger channels d1, d2, d3, ..., interleaved with the source finger channels, provide as many drain channels for the electrolyte directed to the exit slots 27 and 27' (Fig. 4).

The electrolyte is in this way evenly distributed throughout the mass of porous carbon fiber felt electrode 14 with a resulting low pressure drop.

As schematically indicated by the idealized flow arrows in Fig. 4, the electrolyte is practically subjected to flow laterally through a limited segment of carbon felt, effectively refreshing the electrolyte throughout the active electrode surface, on account of the fact that the cooperating interleaved source and drain channelworks provide for a flow path with a negligible pressure drop across the electrolyte flow chamber.

It has been found that with such a channelwork electrode configuration, the carbon felt electrode may occupy the entire depth of the electrolyte flow chamber of the

cell, allowing for the membrane separator to abut directly against the surface of the channelled carbon felt electrode, minimizing ohmic drop in the electrolyte.

The two channelworks may be defined in the carbon felt electrode by cutting it after having been bonded to the bipolar electrode plate 13 or before. Indeed, a carbon felt may be pre-defined by cutting it with a die punch and adhered to a backing sheet to facilitate its handling throughout the bonding process. The backing sheet may ultimately be peeled off the surface of the bonded electrode or removed in any other suitable manner.

Fig. 5 is an enlarged detailed view of the peculiar structure of the composite bipolar plate electrode of the battery of the invention.

Fig. 6 is a cross sectional three-dimensional view of an assembled battery stack.

The bottom end electrode, that in the example shown is constituted by a positive carbon electrode, T+ and the upper terminal electrode T- are set in sturdy end plates of a molded plastic P+ and P-, respectively, which are reinforced by a stress plate of stainless steel SS, onto which act the compression nuts M upon tensioning the tie rods TR.

The use of plastic end plates P+ and P- facilitate the constitution of a perfectly sealed and corrosion proof terminal for electrical connection of the battery in the external circuit.

As shown, the battery of the invention besides being able to be assembled in a horizontal position, it may also be maintained in such a horizontal position during operation.

The orientation of all positive electrodes under the membrane separator of the respective cell and consequently all negative electrodes above the membrane separator, is a preferred orientation.

In fact, during operation a minimum amount of hydrogen evolution of the negative electrodes and of oxygen evolution on the positive electrodes may occasionally and accidentally occur.

According to this preferred orientation, the eventually evolved oxygen will raise  
5 by buoyancy toward the membrane separator and the hydrogen to the base of the negative electrode, both the oxygen and the hydrogen being eventually flown out with the electrolyte.

In this way, a continued contact of oxygen with the base plate of the carbon electrodes which could provoke oxidation (corrosion) of the carbon is minimized.  
10 Moreover, the polymeric membrane that is substantially permeable to hydrogen could permit a certain migration of hydrogen through the membrane, if the cell stack is overturned.

The ability to mount the cell elements and operate the battery in a substantially horizontal orientation instead of in a traditional vertical orientation, has been  
15 found to be advantageous, not only because of the great ease with which the battery is assembled but also in operating the battery, especially in the case of batteries of a particularly large cell area. The vertical orientation reduces mechanical stresses and permits a more efficient construction of the elements and also eases hydraulic sealing problems.

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## CLAIMS

1. Membrane-separated, bipolar multicell electrochemical reactor for half-cell reduction and oxidation reactions in respective positive and negative liquid electrolytes, without gas evolution, including a plurality of alternately disposed bipolar plate electrode elements and ion exchange membrane separator elements, defining a positive electrolyte flow chamber on one side of each membrane and a negative electrolyte flow chamber on the opposite side thereof, sealingly assembled together in a filter-press arrangement between two end electrode elements electrically coupled into an electric circuit functionally including an electrical source forcing a current through the electrochemical reactor or an electrical load absorbing a current from the electrochemical reactor,

said bipolar plate electrode elements and said ion exchange membrane separator elements including a frame portion of an electrically nonconductive and chemically resistant material cooperating with sealing gasket means and having through holes and recesses in coordinated locations forming, upon assembling, ducts for the separate circulation of a negative electrolyte and of a positive electrolyte, cascadedly in all said negative electrolyte flow chambers and in all said positive electrolyte flow chambers, respectively,

characterized in that  
all the frames of said bipolar plate electrode elements and of said ion exchange membrane separator elements have an inner flange portion, recessed from a first planar face of the frame on the opposite side of the other face of the frame having grooves for accommodating O-ring gaskets around pass-through electrolyte-ducting holes and around an outer seal perimeter, accommodating thereon a perimetral portion of the respective bipolar plate electrode or ion exchange membrane separator;

a plurality of retention pins project out of the surface of said flange portion and pass through holes of said perimetral portion of the plate electrode or membrane separator accommodated thereon;

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a retention counterflange of an electrically nonconductive and chemically resistant material having holes coordinated with the positions of said retention pins and functionally mounted over said perimetral portion of plate electrode or membrane separator on said recessed flange portion of the frame and permanently fixed thereon by hot-flattened heads of said retention pins protruding out of said coordinated holes of the counterflange;

pre-assembled bipolar plate electrode elements and membrane separator elements being alternately stackable in horizontal position with said other face of the frames carrying the O-ring gaskets facing upward.

2. The electrochemical reactor of claim 1, characterized in that said other face and said first planar face of each frame portion are provided with a plurality of keying and alignment pins and sockets, respectively, of different shape from each other, preventing the stacking of said bipolar plate electrode elements and of said ion exchange membrane separator elements in an incorrect alternate order and/or in an incorrect orientation.

3. The electrochemical reactor of claim 1, wherein said inner flange portions, said bipolar plate electrodes and said ion exchange membrane separators are rectangular and said frame portions have convex outer sides.

4. The electrochemical reactor of claim 1, wherein the directions of flow of said negative electrolyte and of said positive electrolyte in respective flow chambers along the opposite sides of each ion exchange membrane separator, are opposite to each other.

5. The electrochemical reactor of claim 1, wherein each of said bipolar plate electrode consists of a fluid impervious electrically conductive plate having on opposite faces thereof porous fluid pervious three-dimensional electrode structures in the form of a felt or fabric of carbon fibers bonded in electrical continuity to said electrically conductive plate, the electrolyte entering the

electrode chamber along one side and exiting the chamber from the opposite side; and further characterized in that said porous electrode structure has two distinct comb-shaped channelworks, the finger channels of one channelwork being substantially parallel to each other and interleaved with the substantially parallel finger channels of the other channelwork; a first or source comb-shaped channelwork having a base or manifolding channel running along the side of the chamber through which the electrolyte is fed into the chamber and the second or drain channelwork having its base or manifolding channel running along the opposite side from which the electrolyte exits the chamber;

all finger channels of one channelwork extending from the respective base or manifolding channel and terminating short of reaching the manifolding channel of the other channelwork.

6. The electrochemical reactor of claim 1, wherein the ducts for the separate circulation of each of said negative and positive electrolytes defined by said through holes across the thickness of each frame portion of said bipolar plate electrode elements and of said ion exchange membrane separator elements are defined by two or more holes spaced along one side of the substantially rectangular frame portion.

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## MEMBRANE-SEPARATED, BIPOLAR MULTICELL ELECTROCHEMICAL REACTOR

### ABSTRACT

5 A multicell assembly is constituted by alternately stacking two types of pre-assembled elements: a bipolar electrode holding subassembly and a membrane holding subassembly. The alternate stack of elements is piled over a bottom end element and the stack is terminated by placing over the last membrane holding element a top end electrode element. Each bipolar plate electrode holding element and each ion exchange membrane separator holding element includes a  
10 substantially similar rectangular frame piece, made of an electrically nonconductive and chemically resistant material, typically of molded plastic material, having on its upper (assembly) face grooves for receiving O-ring type gasket means, and having through holes and recesses in coordinated locations disposed along two opposite sides of the rectangular frame forming, upon  
15 completion of the assembling, ducts for the separate circulation of the negative electrolyte and of the positive electrolyte through all the negative electrolyte flow chambers and all positive electrolyte flow chambers, respectively, in cascade. The bipolar reactor does not have inlet and outlet manifolds for the two electrolytes, on the contrary, the electrolytes flow through the respective flow chambers in a  
20 zigzag path, that is essentially in hydraulic series or cascade mode instead than in hydraulic parallel mode. Therefore, by-pass current may only be "driven" by a relatively low voltage difference of about one-cell voltage.

25 Preferably, two orders of parallel flow channels are defined in felt electrodes to minimize pressure drops, all the parallel spaced channels of each order extend from a common orthogonal base channel formed along the respective inlet or outlet side of the chamber and terminate short of reaching the base channel of the other order. Each order defines a comb-shaped flow distributing channelwork the parallel fingers of which interleave with the finger channels of the other order.

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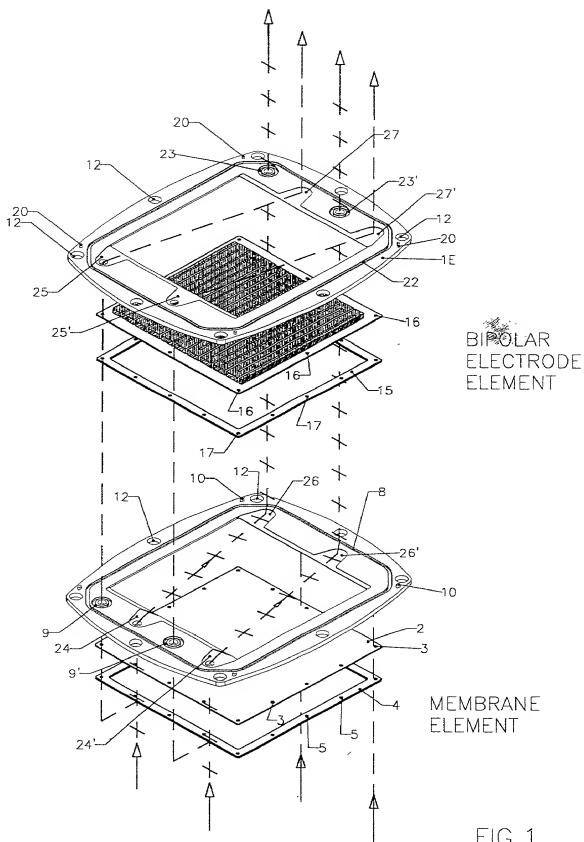


FIG. 1

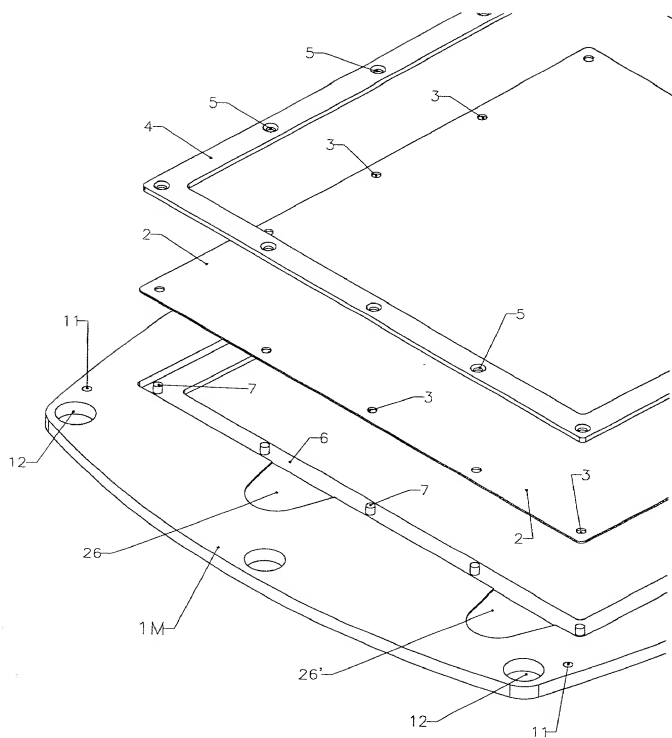


FIG. 2

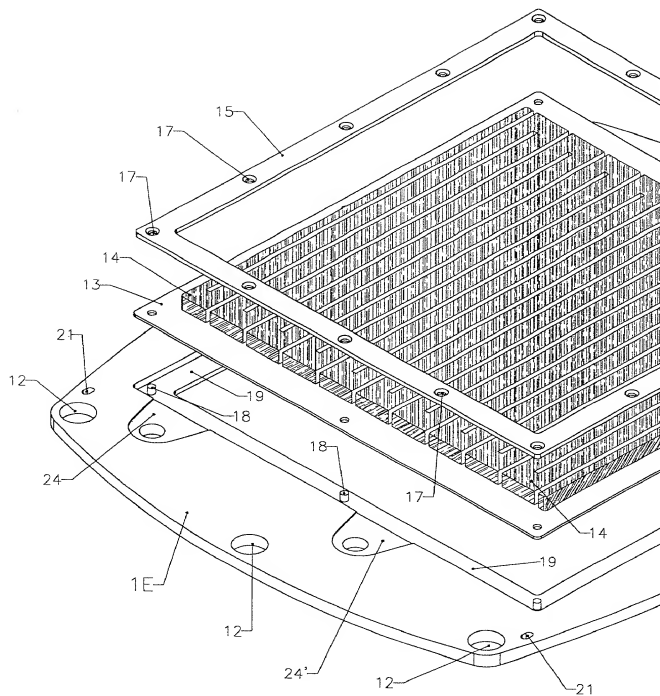


FIG.3

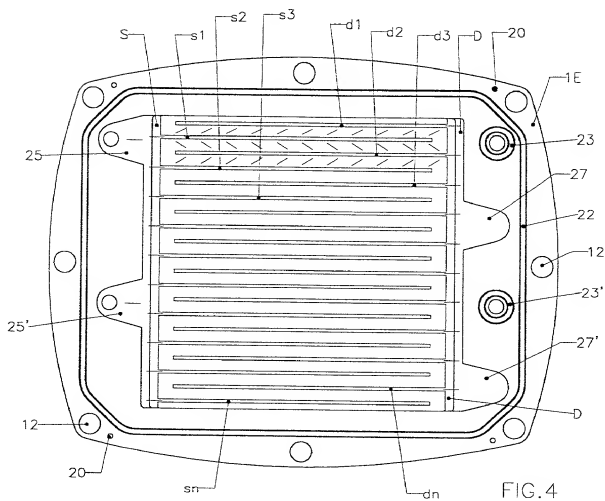


FIG. 4

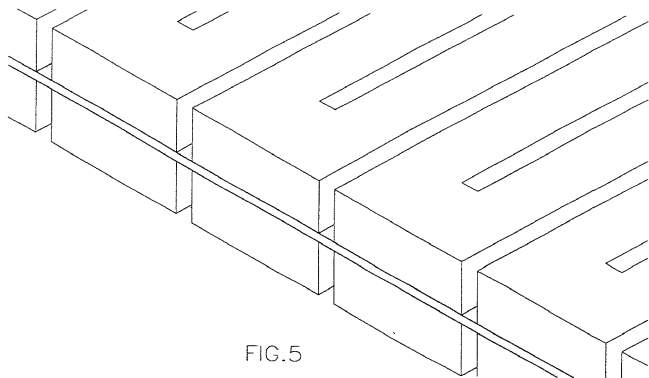


FIG. 5



FIG. 6

## DECLARATION FOR PATENT APPLICATION

0679/00188

As a below-named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

**MEMBRANE-SEPARATED, BIPOLAR MULTICELL ELECTROCHEMICAL REACTOR**

the specification of which: (check one)

☐ is attached hereto.☒ was filed on July 1, 1999, as United States Patent Application Serial No. or PCT International Application Number PCT/US99/00195, and was amended on 19 (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the patentability of this application in accordance with 37 CFR § 1.56(a).

Prior Foreign Application(s): I hereby claim foreign priority benefits under 35 U.S.C. § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate listed below, or § 365(a) of any PCT international application which designated at least one country other than the United States of America, listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Priority Claimed

(Application No.)	(Country)	(Day/Month/Year Filed)	<input type="checkbox"/>	<input type="checkbox"/>
			YES	NO
(Application No.)	(Country)	(Day/Month/Year Filed)	<input type="checkbox"/>	<input type="checkbox"/>
			YES	NO
(Application No.)	(Country)	(Day/Month/Year Filed)	<input type="checkbox"/>	<input type="checkbox"/>
			YES	NO

I hereby claim the benefit under Title 35, United States Code § 119(e) of any United States provisional application(s) listed below:

Application No.

Filing Date

I hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by 35 U.S.C. § 112, first paragraph, I acknowledge the duty to disclose material information as defined in 37 CFR § 1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

(U.S. Application Serial No.)	(U.S. Filing Date)	(Status - patented, pending, abandoned)
(U.S. Application Serial No.)	(U.S. Filing Date)	(Status - patented, pending, abandoned)

I hereby appoint George Vande Sande, Registration No. 17,226; Burton A. Amernick, Registration No. 24,852; Richard Wiener, Registration No. 18,741; Townsend M. Belser, Jr., Registration No. 22,956; Morris List, Registration No. 24,510; Martin Abramson, Registration No. 25,787; George R. Penit, Registration No. 27,369; Elzbieta Chlopecka, Registration No. 32,767; Eric J. Franklin, Registration No. 37,134; Jeffri A. Kaminski, Reg. No. 42,709; and William E. Curry, Registration No. 43,572, my attorneys with full power of substitution and revocation, to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith.

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 18 U.S.C. § 1001 and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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## DECLARATION FOR PATENT APPLICATION

Page 2

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Post Office Address

Full name of sixth joint inventor (if any)

Inventor's Signature

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Full name of eighth joint inventor (if any)

Inventor's Signature

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Citizenship

Post Office Address



## DECLARATION FOR PATENT APPLICATION

0679/00188

As a below-named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

## MEMBRANE-SEPARATED, BIPOLAR MULTICELL ELECTROCHEMICAL REACTOR

the specification of which: (check one)

☐ is attached hereto.☒ was filed on July 1, 1980, as (United States Patent Application Serial No. or PCT International Application Number PCT/US99/01155, and was amended on 19 (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the patentability of this application in accordance with 37 CFR § 1.56(a).

Prior Foreign Application(s): I hereby claim foreign priority benefits under 35 U.S.C. § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate listed below, or § 365(a) of any PCT international application which designated at least one country other than the United States of America, listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Priority Claimed

(Application No.) APR 12 2001 (Country) US  
 (Application No.) 1980 (Country) US  
 (Application No.) 1980 (Country) US

(Day/Month/Year Filed) APR 12 2001  
 (Day/Month/Year Filed) APR 12 2001  
 (Day/Month/Year Filed) APR 12 2001

☐ YES ☐ NO  
☐ YES ☐ NO  
☐ YES ☐ NO

I hereby claim the benefit under Title 35, United States Code § 119(e) of any United States provisional application(s) listed below:

Application No.

Filing Date

I hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s) listed below, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by 35 U.S.C. § 112, first paragraph, I acknowledge the duty to disclose material information as defined in 37 CFR § 1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

(U.S. Application Serial No.)

(U.S. Filing Date)

(Status - patented, pending, abandoned)

(U.S. Application Serial No.)

(U.S. Filing Date)

(Status - patented, pending, abandoned)

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 18 U.S.C. § 1001 and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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# DECLARATION FOR PATENT APPLICATION

Page 2

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